Meandering of monolayer stripes under electromigration

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The dynamics of lines composed of two monatomic steps of opposite sign and periodically spaced on a crystal surface has been investigated under electromigration. It is found that, when adatoms have a diffusion bias parallel to the step edges, lines may become unstable with respect to shape fluctuation, which may lead to the formation of a pattern composed of antiphase serpentinelike lines.

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The control of kinetic instabilities arising during the growth of crystals that exhibit stepped surfaces is of fundamental interest in manufacturing electronic and optoelectronic devices [1–8]. For instance, step bunching, step pairing, and step meandering instabilities have been found to modify the step flow growth regime of crystals. Step bunching has been investigated during molecular beam epitaxy, pulsed laser deposition, or under electromigration [9,10]; it results in the formation of high-density step areas separated by large terraces. In the case of a $SrRuO_3$ film epitaxially grown on a vicinal $SrTIO_3$ substrate by the pulsed laser deposition method, the influence of deposit flux on the different regimes of step flow, step bunching, and island formation has been characterized and the conditions for persistent step flow growth have been determined [7,8]. Step bunching on Si(111) surfaces has also been observed and characterized when an external force induced by heating with a direct electric current is applied to adatoms [11]. The step pairing instability under electromigration has been investigated when the dynamics of steps is nonlocal due to step transparency, this effect being experimentally observed on Si(111) [6]. For metal surfaces, electromigration-induced morphological instability has also been found to play a key role in the degradation of microelectronic devices and the patterning of vicinal surfaces [12,13]. In the case of trains of identical steps, one interesting feature of electromigration is to allow for meandering instabilities with nonzero phase shift between perturbations of two consecutive steps. This instability has been studied as a function of the external force orientation in the linear regime [14] and more recently in the nonlinear regime when diffusion bias is produced by an electric field applied along the line direction [15]. The long-time coarsening dynamics was then characterized in the attachment-detachment-limited and diffusion-limited regimes. In this Brief Report, the effects of electromigration have been investigated on the dynamics of a set of parallel lines periodically distributed on a crystal surface, in the case where the force applied to the adatoms is parallel to the line direction. The resulting morphological changes for lines were then analyzed.

A set of straight lines of width $2h$, height H and spaced out from each other by the same distance $2h$ is considered on the surface of a crystal (see Fig. 1 for the axes). Each line is composed of two monatomic steps of opposite sign labeled steps 1 and 2 and located at $(4n-1)h$ and $(4n+1)h$, respectively. The steps are assumed to be connected through the diffusion field on terraces. An external force $\mathbf{F}=f_y\mathbf{e}_y$ is applied to the adatoms, with f_y, the force magnitude along the $(0y)$ axis, assumed to be constant and positive. Following Stoyanov [9], the resulting drift velocity of adatoms is $D_s\mathbf{F}/k_BT$, with D_s the diffusion rate, k_B the Boltzmann constant, and T the absolute temperature. In the framework of the classical Burton-Cabrera-Franck model [16], the concentration of adatoms, $C_{n,l}$, on the lth terrace of the nth line centered at $x_n=4nh$ satisfies, in the quasistatic limit, the following equation [14]:

$$\nabla^2 c_{n,l} - f_y^* \frac{\partial c_{n,l}}{\partial x} = 0, \tag{1}$$

with ∇^2 the Laplacian, $f_y^*=f_y/k_BT$, and $l=1,2,3$. The evaporation and deposition of adatoms on terraces have been ignored [14]. The boundary conditions at each step of the nth line are written as follows:

$$\mp \mathbf{n}\cdot\mathbf{J}|_\pm = \nu_\pm\,(c_{n,l}-c_{\rm eq})|_\pm, \tag{2}$$

where + and − refer to the lower and upper terraces, respectively, $\mathbf{J}=-D_s\nabla c_{n,l}+D_sf_y^*c_{n,l}\mathbf{e}_y$ is the surface flux of adatoms, $c_{\rm eq}$ is the equilibrium concentration at each step, $\mathbf{n}$ is the unit normal vector pointing to the lower terrace, and $l=1$, 2, or 3 depending at which step and on which terrace Eq. (2) is written. For the sake of simplicity, the Ehrlich-Schwoebel effect [17] has not been considered and the kinetic coefficients $\nu_\pm$ are chosen such that $\nu_+=\nu_-=\nu$, with ν a constant. From mass conservation, the step normal velocity of the pth step of the nth line is written as

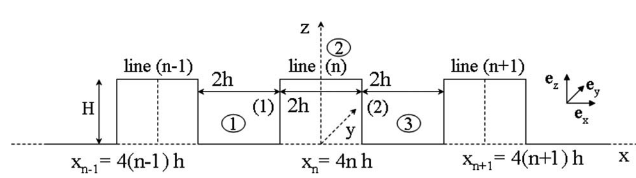

FIG. 1. A periodically spaced distribution of lines is considered on the surface of a crystal. Each line of width $2h$ and height H is composed of two monatomic steps of opposite sign and labeled steps 1 and 2.

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$$v_{n,p} = \Omega \mathbf{n} \cdot (\mathbf{J}|_{-} - \mathbf{J}|_{+}), \tag{3}$$

with $p=1,2$. The equilibrium concentration $c_{\rm eq}$ used in Eq. (2) has been derived from the Gibbs-Thomson relation as follows: $c_{\rm eq}=c^0_{\rm eq}(1+\Gamma\kappa)$, where $c^0_{\rm eq}$ is the equilibrium concentration at the straight step and κ the local curvature of the considered step, taken to be positive for a convex profile. The constant Γ is defined by $\Gamma=\gamma\Omega/k_BT$ with γ the step stiffness and Ω the atomic area of the crystal. To carry out the linear stability analysis, a single Fourier mode of the position of the step (p) of the nth line, $\zeta_{n,p}(y,t)$, has been described in Fourier space, with $p=1,2$. For steps 1 and 2 of the nth line, one takes $\zeta_{n,1}(y,t)=(4n-1)h + e\exp[+iky+in\varphi+\omega(k,\varphi,\xi)t]$+c.c. and $\zeta_{n,2}(y,t)=(4n+1)h + e\exp[+iky+in\varphi+i\xi+\omega(k,\varphi,\xi)t]$+c.c., respectively, where e is the perturbation amplitude, t the time, k the wave number along the step direction, ω the growth rate of the fluctuations, φ the phase factor between two consecutive lines, and ξ the phase shift between the two steps of each line. For symmetry reasons, only in-phase and antiphase configurations for the steps of each line are considered. In the linear regime, one thus takes $\xi=0$ for serpentinelike lines (SLs) and $\xi=\pi$ for pinched lines (PLs). Likewise, the phase shift between two consecutive lines is taken to be $\varphi=0$ or π. The general solution of Eq. (1) is $c_{n,l}(x,y)=c^0_{\rm eq}+c^{(1)}_{n,l}(x,y)$, with $c^{(1)}_{n,l}(x,y)=\exp(+iky)[A^{(1)}_{n,l}\cosh(\lambda x)+B^{(1)}_{n,l}\sinh(\lambda x)]$+c.c. the first-order correction in the perturbation amplitude e to the concentration on terrace l of the nth line, with $c_{n-1,3}=c_{n,1}$, $\lambda=\sqrt{k^2+ikf^*_y}$ and $l=1,2,3$. For each SL and PL configuration, the $A^{(1)}_{n,l}$ and $B^{(1)}_{n,l}$ coefficients have been determined by expanding Eq. (2) up to order 1 in the perturbation amplitude e and matching first-order terms. The heavy but straightforward calculation of these coefficients is not detailed in this Brief Report. In the case of the SL configurations, using Eq. (3), one gets the following growth rates:

$$\begin{aligned}\omega^{\varphi=\pi}_{\rm SL}(k) &= \omega(k,\varphi=\pi,\xi=0)\\ &= 2D_s c^0_{\rm eq}\Omega\\ &\quad\times\frac{id_s f^*_y k\lambda - \Gamma k^2\lambda[\cosh(2h\lambda)+d_s\lambda\sinh(2h\lambda)]}{2d_s\lambda\cosh(2h\lambda)+(1+d_s^2\lambda^2)\sinh(2h\lambda)},\end{aligned} \tag{4}$$

$$\begin{aligned}\omega^{\varphi=0}_{\rm SL}(k) &= \omega(k,\varphi=0,\xi=0)\\ &= 2D_s c^0_{\rm eq}\Omega\frac{-\Gamma k^2\lambda\cosh(h\lambda)}{d_s\lambda\cosh(h\lambda)+\sinh(h\lambda)},\end{aligned} \tag{5}$$

with $d_s=D_s/\nu$ a new kinetic length scale [14]. In the case of the PL configurations, the growth rates are given by

$$\begin{aligned}\omega^{\varphi=\pi}_{\rm PL}(k) &= \omega(k,\varphi=\pi,\xi=\pi)\\ &= 2D_s c^0_{\rm eq}\Omega\\ &\quad\times\frac{-id_s f^*_y k\lambda - \Gamma k^2\lambda[\cosh(2h\lambda)+d_s\lambda\sinh(2h\lambda)]}{2d_s\lambda\cosh(2h\lambda)+(1+d_s^2\lambda^2)\sinh(2h\lambda)},\end{aligned} \tag{6}$$

$$\begin{aligned}\omega^{\varphi=0}_{\rm PL}(k) &= \omega(k,\varphi=0,\xi=\pi)\\ &= 2D_s c^0_{\rm eq}\Omega\frac{-\Gamma k^2\lambda\sinh(h\lambda)}{\cosh(h\lambda)+d_s\lambda\sinh(h\lambda)}.\end{aligned} \tag{7}$$

The stability of SL and PL configurations has been investigated considering the real part of the corresponding growth rates. From Eqs. (5) and (7), it can first be deduced that, since only the smoothing term due to curvature and proportional to Γ is present in the growth rate expressions $\omega^{\varphi=0}_{\rm SL}$ and $\omega^{\varphi=0}_{\rm PL}$, the development in the linear regime of SL or PL in-phase configurations is never favorable. From Eq. (4), apart from negative smoothing terms due to curvature, a new term due to the external force and proportional to if^*_y appears. To determine in which conditions the development of such a SL pattern is favorable or not, the real part of the growth rate $\omega^{\varphi=\pi}_{\rm SL}$ has been expanded as a function of the wave number k in the case of small k values for which the instability is first supposed to appear. The imaginary part of the growth rate giving the modulation of an oscillating contribution in time is not considered. Therefore, assuming $kh\ll 1$ and also $f^*_y \ll k$ in the case of small current intensity, one gets to second order in k

$$\begin{aligned}\mathrm{Re}[\omega^{\varphi=\pi}_{\rm SL}(k)] &= \frac{Dc^0_{\rm eq}\Omega}{3(d_s+h)^2}[(f^*_y)^2(3d_s^3h+6d_s^2h^2+2d_sh^3)\\ &\quad-3\Gamma(d_s+h)]k^2+\cdots,\end{aligned} \tag{8}$$

the higher-order term proportional to k^4 in $\mathrm{Re}(\omega^{\varphi=\pi}_{\rm SL})$, which is too complicated to be written here, being negative. It can be deduced from Eq. (8) that the development of antiphase serpentinelike lines with small wave number k may be favorable under the following condition:

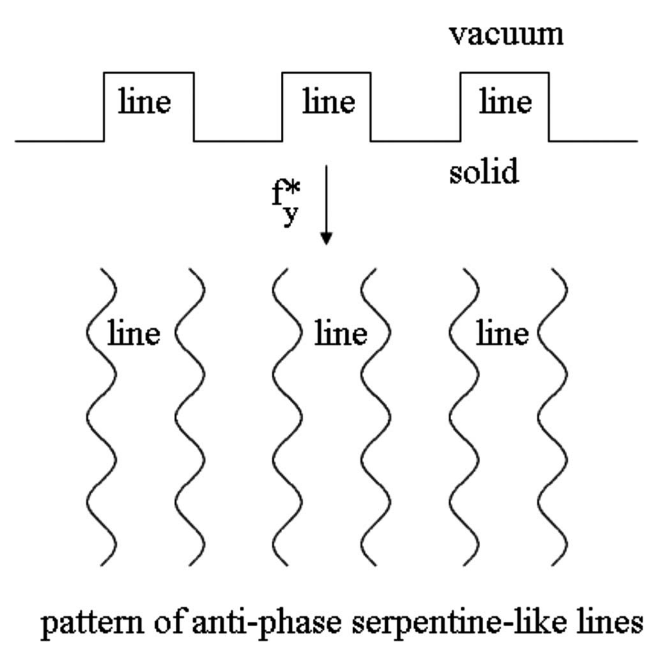

FIG. 2. Electromigration-induced morphological evolution of lines on the crystal surface: formation of a pattern of antiphase serpentinelike lines.

$$\frac{(f_y^*)^2}{\Gamma} \geqslant \frac{(d_s + h)}{d_s^3 h + 2d_s^2 h^2 + \frac{2}{3} d_s h^3}, \quad (9)$$

with the constraints $f_y^* \ll k$ and $kh \ll 1$. When the attachment-detachment kinetics is slow, $d_s \gg h$, taking small values for f_y^* and h, an analytic expression for the critical value $2h_c$ of line thickness above which the instability develops has been determined from Eq. (9) to be

$$h \geqslant h_c = \frac{\Gamma}{(f_y^* d_s)^2}. \quad (10)$$

From the development of $\mathrm{Re}(\omega_{\mathrm{PL}}^{\varphi=\pi})$ as a function of k, it has been found that the term proportional to k^2 is negative while the term proportional to k^4 could be positive depending on the f_y^* value. It can then be concluded that in the early line evolution, when sufficiently long-wavelength perturbations are considered, a pattern composed of antiphase serpentine-like lines as depicted in Fig. 2 may (theoretically) appear.

In conclusion, it is believed that different studies may be considered after this linear stability analysis of the shape of structures composed of steps of opposite sign. From the experimental point of view, it would now be a challenge to find materials and to investigate the experimental conditions for which these line morphological changes can be observed and analyzed. From the theoretical point of view, it would be relevant to investigate the morphological evolution of lines in the nonlinear regime to determine whether or not more complex nonsymmetric patterns than the simple ones that have been considered in this Brief Report (with the same perturbation amplitude e for steps) might emerge under electromigration.

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